Parametric Uncertainty in a Thermal Conductivity Model of Uranium Oxide Light Water Nuclear Reactor Fuel

C. Unal[*1], C.J. Stull[2], B.J. Williams[3]

[*1]Nuclear Engineering and Nonproliferation Division (NEN-DO); [2]Applied Engineering and Technology Division (AET-6); [3]Computer, Computational and Statistical Sciences (CCS-6)

Los Alamos National Laboratory
PO Box 1663, Mail Stop C921

Los Alamos, New Mexico 87545, USA

[*1]cu@lanl.gov; [2]stull@lanl.gov; [3]brianw@lanl.gov

Abstract

In the absence of a well-accepted, mathematical description of how units of material respond to one or more external actions, scientists must often rely upon empirically-derived models to make predictions. These models are often employed to model very complex phenomena with an unfortunate side-effect that intuition, as to the values of particular model parameters, may not serve to guide their use. Given our increased reliance on modeling and simulation to make predictions in the absence of experimental data, it befits the scientific and engineering communities to explore and report upon uncertainty quantification techniques applied to previously adopted (or at least well-accepted) empirical models that are derived from or pertain to substantial experimental data sets. This paper employs Markov Chain Monte Carlo (MCMC) analyses to assess the predictive capability of the empirical thermal conductivity model adopted by the nuclear fuel performance code, FRAPCON-3.4. The effect of uncertain parameters on the ability of the model to predict uranium dioxide conductivity data from open literature sources has been taken into consideration. The results led the authors to question the predictive capability of the FRAPCON model for predicting the thermal conductivities associated with *irradiated* fuel samples.

Keywords

Nuclear; Markov Chain Monte Carlo; Reactor; Parametric; Uncertainty; Thermal Conduction; Uranium Dioxide

Introduction

In the absence of a well-accepted, mathematical description of how units of material respond to one or more external actions (*e.g.* temperature fluctuations), scientists must often rely upon empirically-derived models (*e.g.* polynomials) to make predictions. These predictions may be interpolatory or extrapolatory in nature, but irrespective of this, the predictions are derived from a model that employs little-to-no physics: a potentially dangerous situation if the phenomena modeled are, in reality, complex processes. This situation can be especially disconcerting when limited data sets are available for development of the model or when the model does not generalize well to data sets that become available after its development.

Given our increased reliance on modeling and simulation to make predictions (both interpolatory and extrapolatory) in the absence of experimental data, it befits the scientific community to explore and report upon uncertainty quantification techniques applied to previously adopted (or at least well-accepted) empirical models that derive from, or pertain to, substantial experimental data sets. Such studies, while more applied in nature, provide guidance for conducting studies on codes that are still under development. This paper and the companion paper [Stull, Williams, Unal, 2013] represent a collection of three methodologies aimed at assessing the predictive capability of the empirically-derived thermal conductivity model adopted by the nuclear fuel performance code, FRAPCON-3.4 (referred to as the "FRAPCON model"). The methodologies adopt optimization-based, Bayesian inference based, and info-gap decision theory based approaches to the problem. Due to space limitations, the results from the optimization-based approach are only

briefly mentioned in the companion paper, leaving the companion paper to focus on the info-gap decision theory based approach. This paper then focuses on the results of the Markov Chain Monte Carlo (MCMC) analysis. Following is an overview of the historical development of the FRAPCON model that is considered in this paper and the companion paper.

Thermal Conductivity Model

The ceramic nuclear fuel, UO_2, used in light water reactors is known to be a very poor semiconductor. The heat generated by fission is transferred by thermal conduction primarily by way of lattice vibrations. The dependency of the thermal diffusivity (or thermal conductivity, the inverse of thermal diffusivity) on temperature is traditionally expressed as

$$K_{95} = \frac{1}{A+BT}. \quad (1)$$

In this equation, A represents the effect of phonon-impurity scattering processes, and B represents the effect of phonon-phonon scattering processes [Ronchi et al., 2004], T is temperature and K_{95} is the thermal conductivity at 95% theoretical density. This simple dependency worked well for the thermal conductivity estimation of fresh fuels, as well as of irradiated fuels. There are various excellent papers on the development of an appropriate thermal conductivity model for UO_2. For example, [Ronchi et al., 1999] modified Equation (1) based on new measurements to account for ambipolar and radiation contributions to thermal conductivity. The radiation contribution was found to be small relative to the lattice vibration and ambipolar contributions. As a result, nuclear fuel performance codes like FRAPCON include a thermal conductivity model of the following form:

$$K_{95} = \frac{1}{A+BT} + \frac{E}{T^2}\exp\left(-\frac{F}{T}\right) \quad (2)$$

where $A = 0.0452$, $B = 0.000246$, $E = 3.5E9$, and $F = 16361$. Note that the ambipolar contribution is accounted for by the right-most term and becomes important at temperatures above 1900 K. Fig. 1 presents the available experimental thermal conductivity data associated with fresh fuel samples along with the nominal FRAPCON thermal conductivity model given by Equation (2). It is this model that will be considered in the context of predicting thermal conductivity for fresh fuel samples.

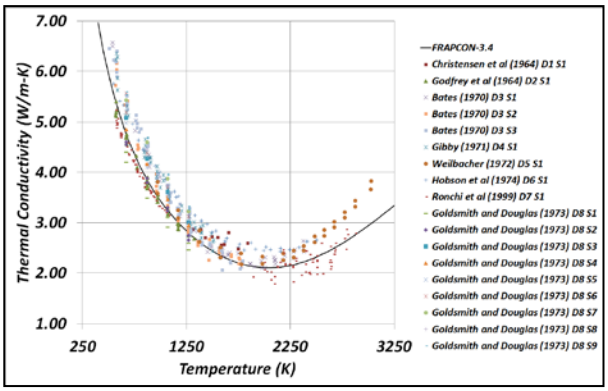

FIG. 1 EXPERIMENTAL TEMPERATURE VS. THERMAL CONDUCTIVITY PLOTS FOR FRESH SAMPLES, WITH NOMINAL FRAPCON MODEL, AS GIVEN BY EQUATION (2)

Several authors have conducted excellent reviews of UO_2 thermal conductivity models (see *e.g.* [Fink, 2002, Lucuta et al., 1996]. These papers have considered various data sets and have subsequently suggested coefficients that are not necessarily consistent (from reference to reference). With new measurements available, these parameter estimates change on almost a yearly basis. For example, FRAPCON-3 and FRAPCON-3.4 implement completely different thermal conductivity models [Lanning, et al., 1997; Geelhood, et al., 2011].

Nevertheless, the measurement of thermal conductivity of UO_2 is not straightforward, and *in situ* measurements during irradiation are not available. Typically, these measurements are initiated by irradiating the fuel at certain irradiation temperatures that may vary in a nuclear core, depending upon the location. The fuel is then cooled and small samples are analyzed in the laboratory by heating the sample to a desired temperature. Note that this measurement process also introduces annealing effects above certain temperatures. Furthermore, the fission process constantly produces defects that affect the correctness of A and B. These findings have led [Ronchi et al., 2004] to suggest an alternative model for the lattice contribution:

$$K_{95} = \frac{1}{A(T_{\text{irr}},T_{\text{ann}},Bu)+B(T_{\text{irr}},T_{\text{ann}},Bu)T'} \quad (3)$$

where A and B are now *functions*, and T_{irr}, T_{ann}, and Bu are the irradiation temperature, annealing temperature, and burnup (as a percentage), respectively. FRAPCON -3 used a thermal conductivity model suggested by the [Lucuta et al., 1996]. FRAPCON-3.4, on the other hand, claims that the thermal conductivity model is based on the expression developed by [Ohira and Itagaki, 1997] for the Nuclear Fuels Industries. This is given as

$$K_{95} = \frac{1}{A+BT+f(Bu)+g(Bu)h(T)} + CT^2 + DT^4 \quad (4)$$

Where

$$f(Bu) = 0.00187Bu \quad (5)$$

accounts for the effect of soluble fission products,

$$g(Bu) = 0.038Bu^{0.28} \quad (6)$$

accounts for the effect of irradiation defects,

$$h(T) = \frac{1}{1+396\exp(-Q/T)} \quad (7)$$

accounts for the effect of recovery of irradiation defects, and $Q = 6380$. Note that the electronic contribution (*i.e.* the ambipolar effects) is formulated differently in this model than in Equation (2). The actual thermal conductivity model implemented in FRAPCON-3.4 is given as

$$K_{95} = \frac{1}{A+BT+f(Bu)+(1-0.9\exp(-0.04Bu))g(Bu)h(T)} + \frac{E}{T^2}\exp\left(-\frac{F}{T}\right). \quad (8)$$

While $f(Bu)$, $g(Bu)$, and $h(T)$ are retained from Equation (4), a new term $\left(1-0.9\exp(-0.04Bu)\right)$ is introduced in FRAPCON-3.4. There is no good physics argument provided in the FRAPCON manual as to why this new term is introduced. Furthermore, this model also drops the form of the electronic contribution suggested by [Ohira and Itagaki, 1997] and instead adapts the form suggested by [Lucuta et al., 1996], [Fink, 2000] and [Ronchi et al., 2004]. Fig. 2 presents the available experimental thermal conductivity data[1] associated with irradiated fuel samples along with the nominal FRAPCON thermal conductivity model given by Equation (8). It is this model that will be considered in the context of predicting thermal conductivity for irradiated fuel samples.

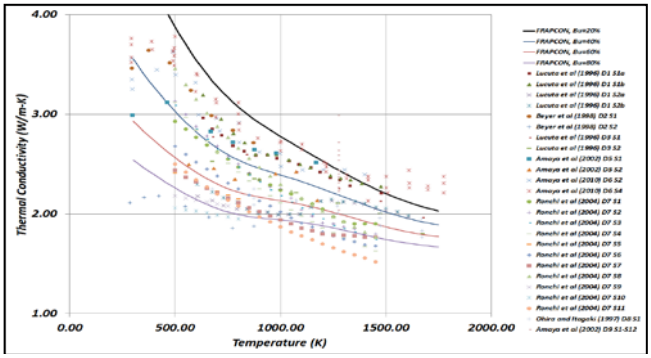

FIG. 2 EXPERIMENTAL TEMPERATURE VS. THERMAL CONDUCTIVITY PLOTS FOR IRRADIATED FUEL SAMPLES WITH NOMINAL FRAPCON MODEL, AS GIVEN BY EQUATION (8)

[1] The data sets labeled "Amaya et al., 2002, D5 S1," "Amaya et al., 2002, D5 S2", "Amaya et al., 2010, D6 S2," and "Amaya et al., 2010, D6 S4" in Fig. 2 are not included in the proceeding analyses due to inconsistencies identified by the authors.

TABLE 1 VALUES OF THERMAL CONDUCTIVITY MODEL PARAMETERS FROM DIFFERENT REFERENCES

Parameter Name	FRAPCON3	FRAPCON3.4	Ohira and Itagaki	Lucuta et al.	Ronchi et al.	Fink
A (m-K/W)	0.0375	0.0452	0.0452	0.0375	0.06548	0.0754
B (m-K/W/K)	2.165E-4	2.46E-4	2.46E-4	2.165E-4	0.23533	0.1769
E (W-K/m)	4.715E9	3.5E9	-	4.715E9	6400*	6400*
F (K)	16361	16361	-	16361	16.35	16.35
C (W/m-K/K²)	-	-	-5.47E-9	-	-	-
D (W/m-K/K⁴)	-	-	2.29E-14	-	-	-
*divided by $T^{5/2}$ not T^2						

There is no a good explanation in [Ohira and Itagaki, 1997] why the functions f, g, and h should be in the form of $f + gh$. Their dependencies on the coefficients A and B and their potential correlations are also not known. Table 1 presents the coefficients of A, B, E, and F recommended by different authors, indicating a large degree of variation.

The fuel companies may also have different coefficients specific to their fuels. It is expected that the coefficients A, B, E, and F are calibrated from fresh fuel data first and irradiation effects are introduced by examining trends in the irradiated data. In this research, the authors collected as much data from open literature as could be found, in order to assess the model parameters, independent of model developers. It is asserted by the authors that the irradiation effects should be included in the lattice term as suggested by [Ronchi et al., 2004]. Instead, previous authors have introduced additive terms in the denominator of the thermal conductivity equation.

In this paper, the authors consider the thermal conductivity models in FRAPCON-3.4 (*i.e.* Equations (2) and (8) for the fresh and irradiated fuel samples, respectively) and attempt to understand the correlation issues between A and B from Equation (2) and f, g, h, and the extra term from Equation (8).

Markov Chain Monte Carlo Analyses

In parametric Bayesian analysis, observed data $\boldsymbol{y}$ are sampled from a distribution $g(\boldsymbol{y}|\boldsymbol{\theta})$, where g is a specified family of distributions (*e.g.* Gaussian) and $\boldsymbol{\theta}$ is a collection of parameters having unknown values. The family of distributions g is often determined by the nature of experimental errors. For example, a particular fuel conductivity dataset could be modeled as follows,

$$\boldsymbol{y} = \boldsymbol{\eta}(\boldsymbol{\theta}) + \boldsymbol{\varepsilon} \quad (9)$$

where $\boldsymbol{\eta}(\boldsymbol{\theta})$ is the vector of conductivity model outputs evaluated at model parameters $\boldsymbol{\theta}$ and corresponding to the conditions producing $\boldsymbol{y}$, and $\boldsymbol{\varepsilon}$ is the vector of observational errors. These errors are often assumed to be independent, zero-mean, Gaussian with specified standard deviations. In this scenario, pursued in the subsequent analyses, the family g is multivariate Gaussian having mean $\boldsymbol{\eta}(\boldsymbol{\theta})$ and specified standard deviations.

In the following discussion, the sampling distribution g viewed as a function of unknown parameters $\boldsymbol{\theta}$ (for given data values $\boldsymbol{y}$) will be referred to as the *likelihood function*, $L(\boldsymbol{\theta}|\boldsymbol{y}) \equiv g(\boldsymbol{y}|\boldsymbol{\theta})$. For varying $\boldsymbol{\theta}$, higher likelihood values indicate parameter settings for which the conductivity model output exhibits greater consistency with the observed conductivity data $\boldsymbol{y}$. To complete the Bayesian framework, initial (prior) uncertainty in the values of $\boldsymbol{\theta}$ is represented by a distribution π, referred to as the *prior distribution*. In the subsequent analyses, π is assumed to be a Uniform distribution on pre-specified ranges for each model parameter presented in Equations (2) and (8). Given observed conductivity data $\boldsymbol{y}$, inference about $\boldsymbol{\theta}$ is based entirely on the *posterior distribution*, given by

$$\pi(\boldsymbol{\theta}|\boldsymbol{y}) = \frac{L(\boldsymbol{\theta}|\boldsymbol{y})\boldsymbol{\pi}(\boldsymbol{\theta})}{\int L(\boldsymbol{\theta}|\boldsymbol{y})\boldsymbol{\pi}(\boldsymbol{\theta})d\boldsymbol{\theta}}. \quad (10)$$

In words, the posterior distribution utilizes the conductivity data $\boldsymbol{y}$ to update the prior uncertainty about $\boldsymbol{\theta}$, resulting in new uncertainty quantification for $\boldsymbol{\theta}$ that incorporates information from both prior knowledge π and observational data $\boldsymbol{y}$.

In the analyses of this section, the posterior normalizing constant $\int L(\boldsymbol{\theta}|\boldsymbol{y})\boldsymbol{\pi}(\boldsymbol{\theta})d\boldsymbol{\theta}$ is not computable in closed form, as the conductivity models are non-linear. Therefore, exact posterior calculations are not possible. Fortunately, technology exists for generating samples $\boldsymbol{\theta}_i$ from the posterior distribution $\pi(\boldsymbol{\theta}|\boldsymbol{y})$. The methodology employed is referred to as *Markov Chain Monte Carlo* (MCMC) sampling, which involves establishing the target distribution $\pi(\boldsymbol{\theta}|\boldsymbol{y})$ as the stationary distribution of a Markov Chain. The following sampling algorithm used in the subsequent analyses is one of many possibilities that can accomplish this goal:

1. Set the iteration counter $i = 1$ and select a starting value for $\boldsymbol{\theta}$, denoted $\boldsymbol{\theta}^{(0)}$.
2. Suppose that $\boldsymbol{\theta}$ has m elements, of which the first $(j-1)$ have already been updated. The j^{th} parameter is updated as follows. Let $\boldsymbol{\theta}_c = \left(\theta_1^{(i)}, \ldots, \theta_{j-1}^{(i)}, \theta_j^{(i-1)}, \theta_{j+1}^{(i-1)}, \ldots, \theta_m^{(i-1)}\right)$ be the current value of $\boldsymbol{\theta}$, and propose a new value for θ_j (denoted θ_j^*) by sampling from the specified distribution $q_j\left(\theta|\theta_j^{(i-1)}\right)$. Taking $\boldsymbol{\theta}_* = \left(\theta_1^{(i)}, \ldots, \theta_{j-1}^{(i)}, \theta_j^*, \theta_{j+1}^{(i-1)}, \ldots, \theta_m^{(i-1)}\right)$ we set $\theta_j^{(i)} = \theta_j^*$ with probability $\min\left\{1, \frac{L(\boldsymbol{\theta}_*|\boldsymbol{y})\boldsymbol{\pi}(\boldsymbol{\theta}_*)q_j(\boldsymbol{\theta}_c|\boldsymbol{\theta}_*)}{L(\boldsymbol{\theta}_c|\boldsymbol{y})\boldsymbol{\pi}(\boldsymbol{\theta}_c)q_j(\boldsymbol{\theta}_*|\boldsymbol{\theta}_c)}\right\}$; otherwise set $\theta_j^{(i)} = \theta_j^{(i-1)}$. Once the m^{th} parameter has been updated in this fashion, the i^{th} iteration has been completed and the resulting sample is denoted as $\boldsymbol{\theta}_i$.
3. Repeat the previous step n times, resulting in n samples $\boldsymbol{\theta}_1, \ldots, \boldsymbol{\theta}_n$.

An initial subset of iterations $\boldsymbol{\theta}_i$ are discarded as burn-in samples, as these iterations are required for the Markov chain to "forget" its starting value $\boldsymbol{\theta}^{(0)}$ and converge to its stationary distribution (*i.e.* the posterior). It is desirable to choose a $\boldsymbol{\theta}^{(0)}$ having reasonably high posterior probability. In our analyses, $\boldsymbol{\theta}^{(0)}$ is set to be the nominal values of the model parameters (see Equations (2) and (8)). The proposal distribution $q_j(\theta|\theta_c)$ is taken to be uniform on the interval $\left(\theta_c - s_j/2, \theta_c + s_j/2\right)$. The interval width s_j is chosen adaptively during the burn-in period of the MCMC algorithm. Specifically, the algorithm of [Graves, 2011] is used: a specified nominal value of s_j is expanded and contracted dyadically by a specified number of levels. During burn-in, the algorithm repeatedly cycles through each resulting level, collecting acceptance statistics. At the conclusion of burn-in, the acceptance statistics are used to fit a logistic regression model of acceptance probability versus interval width. The interval width s_j is estimated by choosing the value corresponding to a targeted acceptance rate, approximately 40% in the subsequent analyses. The estimated interval widths for each parameter are then used in the above MCMC algorithm post burn-in to generate samples of $\boldsymbol{\theta}$ from the posterior distribution $\pi(\boldsymbol{\theta}|\boldsymbol{y})$. For post-burn-in samples $\boldsymbol{\theta}_{b+1}, \ldots, \boldsymbol{\theta}_n$, posterior conductivity model predictions for any specified fuel conditions are obtained by computing $\boldsymbol{\eta}(\boldsymbol{\theta}_{b+1}), \ldots, \boldsymbol{\eta}(\boldsymbol{\theta}_n)$. Further information on technical and practical considerations for MCMC and its implementation can be found in [Gilks, 1996].

Let $\boldsymbol{\theta}_f$ and $\boldsymbol{\theta}_r$ denote the parameters in the conductivity models of fresh (*i.e.* Equation (2)) and irradiated (*i.e.* Equation (8)) fuel samples, respectively. Here $\boldsymbol{\theta}_r = \left(\boldsymbol{\theta}_f, \boldsymbol{\theta}_{r-f}\right)$, where $\boldsymbol{\theta}_{r-f}$ represents parameters in the irradiated fuel conductivity model that are not in common with the fresh fuel

conductivity model. Three analyses will be conducted: (a) Calibration of $\boldsymbol{\theta}_f$ to fresh fuel conductivity data only, (b) Calibration of $\boldsymbol{\theta}_r$ to irradiated fuel conductivity data only, and (c) Simultaneous calibration of $\boldsymbol{\theta}_f$ and $\boldsymbol{\theta}_{r-f}$ to fresh and irradiated fuel data. Note that (c) facilitates discovery of parameter settings $\boldsymbol{\theta}_f$ and $\boldsymbol{\theta}_{r-f}$, with uncertainty quantification, that are consistent with all available experimental data. The likelihood functions for analyses (a), (b) and (c) are given by Equations (11), (12), (13), respectively, as

$$L_f\left(\boldsymbol{\theta}_f \middle| \boldsymbol{y}_1^f, \dots, \boldsymbol{y}_{n_f}^f\right) = \prod_{i=1}^{n_f} L_f\left(\boldsymbol{\theta}_f \middle| \boldsymbol{y}_i^f\right), \quad (11)$$

$$L_r\left(\boldsymbol{\theta}_r \middle| \boldsymbol{y}_1^r, \dots, \boldsymbol{y}_{n_r}^r\right) = \prod_{i=1}^{n_r} L_r\left(\boldsymbol{\theta}_r \middle| \boldsymbol{y}_i^r\right), \quad (12)$$

$$L_{fr}\left(\boldsymbol{\theta}_f, \boldsymbol{\theta}_{r-f} \middle| \boldsymbol{y}_1^f, \dots, \boldsymbol{y}_{n_f}^f, \boldsymbol{y}_1^r, \dots, \boldsymbol{y}_{n_r}^r\right) = \prod_{i=1}^{n_f} L_f\left(\boldsymbol{\theta}_f \middle| \boldsymbol{y}_i^f\right) \prod_{i=1}^{n_r} L_r\left(\boldsymbol{\theta}_f, \boldsymbol{\theta}_{r-f} \middle| \boldsymbol{y}_i^r\right), \quad (13)$$

where n_f and n_r are the numbers of fresh and irradiated fuel conductivity datasets, respectively. For each dataset, it is assumed that the specified standard deviations in the $\boldsymbol{\varepsilon}$ error model are equal to one-third of 10% of the observed data values.

Results

The results presented in this section are organized around three sub-sections that pertain to three combinations of data sets and model parameters. Section 1) pertains to thermal conductivity data associated with fresh fuel samples, which is analyzed using the FRAPCON-3.4 thermal conductivity model given by Equation (2). Section 2) then pertains to thermal conductivity data associated with irradiated fuel samples, which is analyzed using the FRAPCON-3.4 thermal conductivity model given by Equation (8). Section 3) pertains to the thermal conductivity data associated with both the fresh and irradiated fuel samples in a single MCMC analysis, using both FRAPCON-3.4 thermal conductivity models given by Equations (2) and (8).

The analyses were performed for two different sets of data, corresponding to fresh fuel conditions and irradiated fuel conditions. For each set of data, a fixed number of data sets were used. The fresh fuel data set included eighteen data sets from eight researchers; the irradiated fuel data set included twenty-one data sets from five researchers.

The MCMC analysis adopted 15,500 burn-in samples and 100,000 production runs, where it is noted that an emulator was not required, as the computational burden imposed by the FRAPCON conductivity model equations was minimal, especially when making use of the vectorized computation features available within MATLAB. The nominal values assigned to the uncertain parameters are obtained from the FRAPCON manual, and an allowable range of +/-50% has been utilized, primarily to maintain consistency between the UQ methodologies explored in the companion paper [Stull, Williams, Unal, 2013].

1) Data/Model from Fresh Fuel Samples

Table 2 presents the mean values of the parameters generated by MCMC Analysis (a), in which the data/model associated with the fresh fuel samples only are considered. The third parameter (E) varies considerably from the nominal value, while the remaining parameters vary by what may reasonably be considered an acceptable level (*i.e.* +/-10%). Table 3 does show that the mean values of the parameters produce an improvement in the overall predictive capability of the model, vis-à-vis a reduction in the value of the $E_{\mathrm{RMS}}(\boldsymbol{\theta})$, given by

$$E_{\mathrm{RMS}}(\boldsymbol{\theta}) = \sum_{i=1}^{n}\left(\frac{1}{m_i}\sum_{j=1}^{m_i}\left(K_{i,j}^{\exp} - K_{i,j}^{\mathrm{mod}}(\boldsymbol{\theta})\right)^2\right)^{\frac{1}{2}}. \quad (14)$$

n corresponds to the number of data sets available for the analysis, and m_i corresponds to the number of data points available within the ith data set. Although the mean parameter values from the MCMC analyses are reported in this paper, the authors note that this is done primarily for comparison purposes with other methodologies. The primary purpose of the MCMC analyses reported upon herein is to permit examination of the posterior distributions of the parameters.

TABLE 2 MEAN PARAMETER VALUES FROM MCMC ANALYSIS (A), ALLOWING 50% DEVIATION FROM THE NOMINAL PARAMETER

Parameter Name	Nominal Value	MCMC Mean Value	Percent Difference	Standard Deviation	Percent of Mean
A (m-K/W)	0.0452	0.04696	3.9%	0.00149	3.2%
B (m-K/W/K)	0.000246	0.000236	4.1%	0.0000016	0.7%
E (W-K/m)	3.5E9	5.075E9	45.0%	0.1605E9	3.2%
F (K)	16361	17513.2	7.0%	94.7	0.5%

Arguably, the most attractive products of MCMC analyses that are not offered by many optimization -based analyses are the estimates of the posterior distributions associated with each parameter. For MCMC Analysis (a), the posteriors (univariate and bivariate) associated with the four parameters are presented in Fig. 3 where the order of the parameters is left-to-right / top-to-bottom (*e.g.*

posteriors associated with parameter A are on the first row/column, and so on).

TABLE 3 OBJECTIVE FUNCTION EVALUATIONS ASSOCIATED WITH THE MEAN PARAMETER VALUES FROM MCMC ANALYSIS (FRESH FUEL PARAMETERS / DATA ONLY)

Analysis	$E_{\mathrm{RMS}}(\boldsymbol{\theta})$	Relative Improvement (%)
Nominal	5.22	-
MCMC (50%)	4.43	15.1%

There are two primary results of interest from Fig. 3. The first is with regard to the third (E) parameter, in that the univariate posterior distribution indicates that a more suitable value may lie outside of the prescribed +/-50%. The second result of interest pertains to the presence of a negative correlation between the first (A) and second (B) parameters and the presence of a positive correlation between the third (E) and fourth (F) parameters. The format of Equation (2) explains the presence of these relationships, but of interest is whether these relationships remain present in MCMC Analyses (b) and (c). A reasonable expectation is that the additional parameters associated with Equation (8) will reduce the influences of the first through fourth parameters on the conductivity, thereby diminishing the correlations. However, an equally reasonable expectation is that the combination of the fresh and irradiated data sets (*i.e.* MCMC Analysis (c)) will cause variances on the posteriors to reduce, possibly enhancing the correlations.

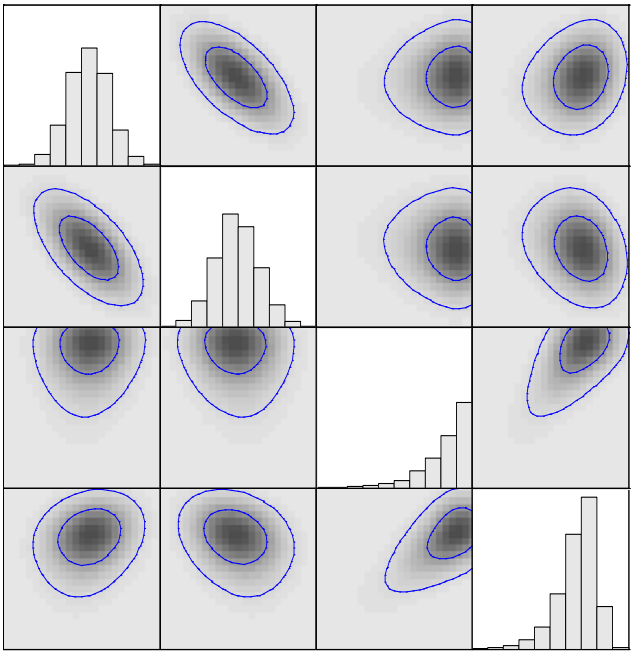

FIG. 3 ESTIMATES OF UNIVARATE (DIAGONAL) AND BIVARIATE (OFF-DIAGONAL) MARGINAL POSTERIOR DISTRIBUTIONS FOR MCMC ANALYSIS (A)

2) *Data/Model from Irradiated Fuel Samples*

TABLE 4 MEAN PARAMETER VALUES FROM MCMC ANALYSIS (B), ALLOWING 50% DEVIATION FROM THE NOMINAL PARAMETER VALUES

Parameter Name	Nominal Value	MCMC Mean Value	Percent Difference	Standard Deviation	Percent of Mean
A (m-K/W)	0.0452	0.06433	42.3%	0.00334	5.2%
B (m-K/W/K)	0.000246	0.0003230	31.3%	0.0000040	1.2%
E (W-K/m)	3.50E+09	1.950E+09	44.3%	0.207E+09	10.6%
F (K)	16361	12079.9	26.2%	192.5	1.6%
f(Bu)	0.00187	0.00095	49.5%	0.000010	1.0%
0.9 Factor	0.9	0.60	33.3%	0.15	24.9%
0.04 Factor	0.04	0.042	5.0%	0.013	30.5%
$g(Bu)_A$	0.038	0.0373	1.8%	0.0089	23.7%
$g(Bu)_B$	0.28	0.289	3.2%	0.053	18.5%
h(T)	396	350.2	11.6%	107.8	30.8%
Q	6380	4418.9	30.7%	253.5	5.7%

TABLE 5 OBJECTIVE FUNCTION EVALUATIONS ASSOCIATED WITH THE MEAN PARAMETER VALUES FROM THE MCMC ANALYSIS (IRRADIATED FUEL PARAMETERS / DATA ONLY)

Analysis	$E_{\mathrm{RMS}}(\boldsymbol{\theta})$	Relative Improvement (%)
Nominal	7.19	-
MCMC (50%)	9.41	-30.9%

Table 4 presents mean values for eight of the eleven parameters that deviate from the nominal values by more than +/-10%, with six of those deviating by more than +/-30%. More disconcerting than the results of Table 4, however, is the substantial reduction in agreement of the model with the experimental data, as presented by the *increased* value of the $E_{\mathrm{RMS}}(\boldsymbol{\theta})$ in Table 5.

Fig. 4 presents the posteriors associated with the MCMC analysis. Of concern are the rather dramatic changes of the posteriors associated with the first, third, and fourth parameters, as compared to those presented in Fig. 3. The posterior associated with the first parameter changes from a Gaussian-like distribution to an Exponential-like distribution, and those associated with the third and fourth parameters change from "favoring" the upper to the lower bound. This is a particularly disturbing result, as it indicates that the two data sets associated with the fresh and irradiated fuel samples may not support the use of common values for the first four parameters of the FRAPCON model. In the very least, the independent calibrations of the model against data sets associated with fresh and irradiated fuel samples should be conducted with caution.

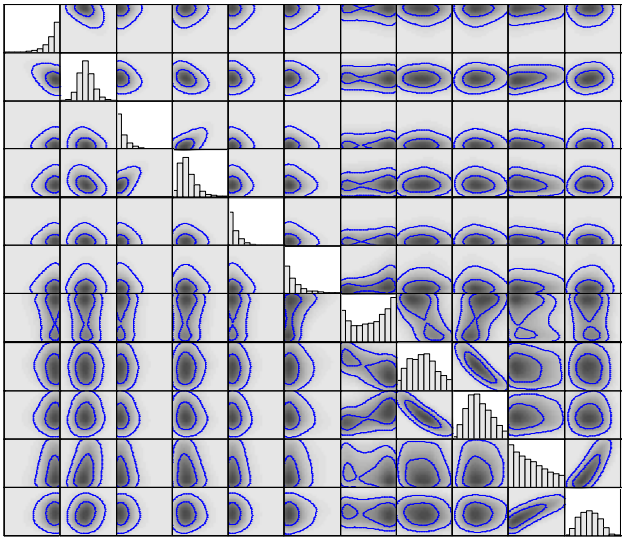

FIG. 4 ESTIMATES OF UNIVARIATE (DIAGONAL) AND BIVARIATE (OFF-DIAGONAL) MARGINAL POSTERIOR DISTRIBUTIONS FOR MCMC ANALYSIS (B)

Of additional concern is the fact that the seventh through eleventh parameters exhibit marginal posteriors that, for the most part, span the entire permissible ranges of the parameter values. Nonetheless, there are two strong correlations between the eighth $(g(Bu)_{\mathrm{A}})$ and ninth $(g(Bu)_{\mathrm{B}})$ parameters (negative) and between the tenth $(h(t))$ and eleventh (Q) parameters (positive). Again, these correlations are explained by Equation (8), but it is again unknown whether or not these will persist in MCMC Analysis (c); for example, the correlation between the first (A) and second (B) parameters is negligible in this analysis, where it was relatively strong in the previous analysis.

The result only goes to confirm that employing data associated with irradiated fuel samples alone for calibration of the FRAPCON model does not yield a predictive model.

3) Data/Model from Fresh and Irradiated Fuel Samples

The attractiveness of this analysis (versus those reported in the companion paper [Stull, Williams, Unal, 2013]) is that it represents the only analysis that *simultaneously* considers all of the data/models. This is particularly interesting in that it allows for the comparison of the posteriors generated by this analysis with those of the previous two analyses, in which only the data/models for the fresh or irradiated fuel samples were considered separately. The first of these two comparisons is presented in Fig. 5, where it is seen that the posteriors are nearly identical between MCMC Analyses (a) and (c). Likewise, the mean parameter values derived from both analyses are also nearly identical (see Table 2 and Table 8), which leads to nearly identical values reported for the $E_{\mathrm{RMS}}(\boldsymbol{\theta})$ associated with the fresh fuel samples (see Table 2 and Table 7).

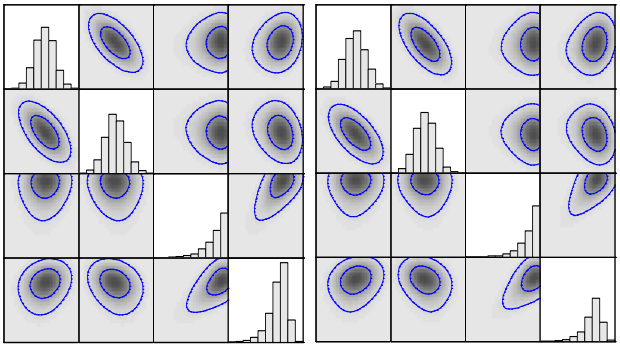

FIG. 5 COMPARISON OF UNIVARIATE (DIAGONAL) AND BIVARIATE (OFF-DIAGONAL) MARGINAL POSTERIOR DISTRIBUTIONS FROM MCMC ANALYSIS (A) AND (C)

TABLE 6 OBJECTIVE FUNCTION EVALUATIONS ASSOCIATED WITH THE MEAN PARAMETER VALUES FROM THE MCMC ANALYSIS (FRESH FUEL PARAMETERS, FRESH AND IRRADIATED DATA)

Analysis	$E_{\mathrm{RMS}}(\boldsymbol{\theta})$	Relative Improvement (%)
Nominal	5.22	-
MCMC (50%)	4.46	14.5%

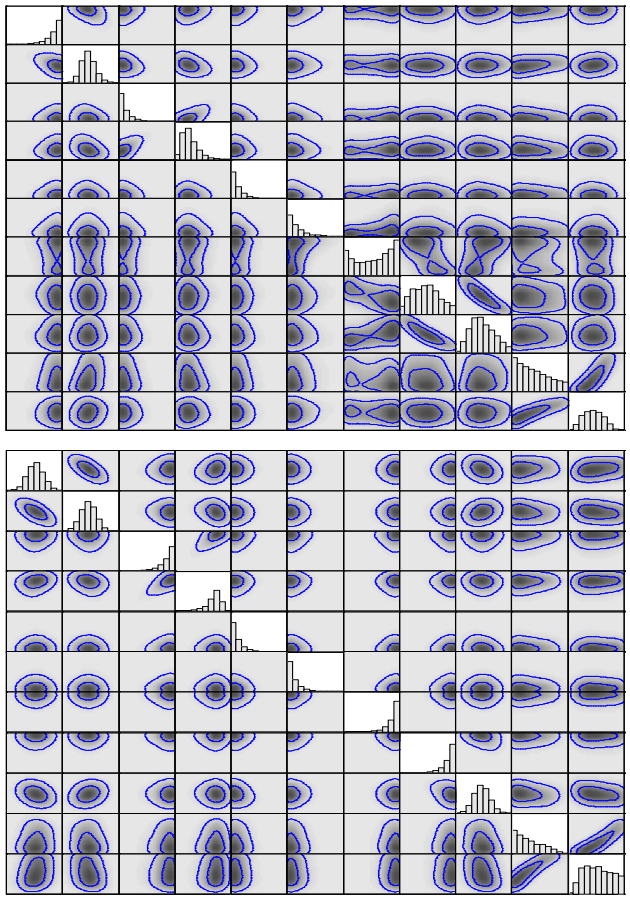

FIG. 6 COMPARISON OF UNIVARIATE (DIAGONAL) AND BIVARIATE (OFF-DIAGONAL) MARGINAL POSTERIOR DISTRIBUTIONS FROM MCMC ANALYSES (A) AND (C)

Of more interest, however, is the comparison of the posteriors associated with *uncommon* parameters (*i.e.* the fifth through eleventh parameters). For this, the authors turn to comparing the posteriors resulting from MCMC Analyses (b) and (c), which are presented in Fig. 6. Most obvious is the fact that variances on the posteriors associated with the sixth through eleventh parameters are significantly reduced. That is, the inclusion of data associated with fresh fuel samples in this combined analysis has a dramatic effect on the values of parameters that *do not* pertain to fresh fuel samples. Recalling the comparison plots between experimental data and nominal model predictions for both fresh and irradiated fuel samples (see Fig. 1 and Fig. 2, respectively), this is not, in fact, an unexpected result. As noted previously, the nominal model associated with the fresh fuel samples offered a reasonably accurate representation of the available data (see Fig. 1). Analogously, MCMC Analysis (a) generated posteriors that were significantly more informative than the assumed Uniform priors. Turning now to the nominal model associated with the irradiated fuel samples, it was noted that the fit of the model to the data was very poor, which translated into the rather uninformative posteriors generated by MCMC Analysis (b).

TABLE 7 OBJECTIVE FUNCTION EVALUATIONS ASSOCIATED WITH THE MEAN PARAMETER VALUES FROM THE MCMC ANALYSIS (IRRADIATED FUEL PARAMETERS, FRESH AND IRRADIATED DATA)

Analysis	$E_{\mathrm{RMS}}(\boldsymbol{\theta})$	Relative Improvement (%)
Nominal	7.19	-
MCMC (50%)	7.80	-8.5%

TABLE 8 MEAN PARAMETER VALUES FROM MCMC ANALYSIS (C), ALLOWING 50% DEVIATION FROM THE NOMINAL PARAMETER VALUES

Parameter Name	Nominal Value	MCMC Mean Value	Percent Difference	Standard Deviation	Percent of Mean
A (m-K/W)	0.0452	0.04997	10.6%	0.00159	3.2%
B (m-K/W/K)	0.000246	0.000233	5.3%	0.0000017	0.7%
E (W-K/m)	3.50E+09	5.078E+09	45.1%	0.166E+09	3.3%
F (K)	16361	17540.6	7.2%	97.9	0.6%
f(Bu)	0.00187	0.000942	49.6%	0.000007	0.7%
0.9 Factor	0.9	0.46	48.9%	0.01	1.4%
0.04 Factor	0.04	0.060	50.0%	0.0004	0.7%
$g(Bu)_A$	0.038	0.0567	49.2%	0.0003	0.5%
$g(Bu)_B$	0.28	0.238	15.0%	0.003	1.1%
h(T)	396	315.9	20.2%	88.6	28.1%
Q	6380	8806.9	38.0%	396.2	4.5%

Returning to MCMC Analysis (c), the previous two MCMC analyses essentially explain the changes seen in the posteriors associated with the sixth through eleventh parameters. The uninformed posteriors associated with MCMC Analysis (b) do not indicate strong tendencies for particular parameter values. So, when the relatively informed posteriors associated with MCMC Analysis (a) *do* indicate such tendencies, the remaining seven, uninformed parameters that are not common between the data/models must "fit" into this new scenario, in which the first four parameters are effectively fixed. That said, these tightened posteriors and in particular, their means (see Table 8), are far from the associated nominal parameter values, which serves to reinforce our earlier conclusion regarding the questionable predictive capability of the FRAPCON model.

Conclusions

This paper represents an application of Markov Chain Monte Carlo (MCMC) analysis to assess the effect of parametric uncertainty in the thermal conductivity model used to make predictions of fuel performance in light water reactors. The predictive capability of the empirical thermal conductivity model adopted by the nuclear fuel performance code, FRAPCON-3.4 is assessed. The use of info-gap decision theory is discussed in the companion paper [Stull, Williams, Unal, 2013]. The genetic algorithm optimization was also used but was not discussed in this paper. While much insight was gained as to the behavior of the models with respect to the available data, the primary conclusion from the genetic algorithm optimization was that the thermal conductivities associated with the fresh fuel samples could *not* be accurately predicted when employing the data/model from the irradiated fuel samples.

This result was confirmed and expounded upon by the second methodology that adopted a Bayesian approach to infer posterior probabilities of the parameters by way of MCMC analyses. The unique facet of this approach was that it considered the *simultaneous* calibration of the two models to all of the available data, which served to confirm the hypothesis that independent calibration of the models/data should be done with caution, especially when considering the model/data associated with the irradiated fuel samples. The issues associated with this model (*i.e.* Equation (8)) lead the authors to question whether or not the model is of the correct mathematical form. As noted, many authors have proposed variants of Equation (8), all of which have fit

the available data to one degree or another. Therefore, future efforts will focus on addressing this issue, possibly by proposing a model that departs from Equation (8), so as to better predict the overall behavior exhibited by the available data.

It is noted that the data set used in the FRAPCON assessment represents a much smaller data set than what was used herein. Data were collected from the open literature, and includes data given in the FRAPCON manual. However, the data set employed for this study is almost twice the size of that used in the FRAPCON assessment, especially for irradiated fuel. Some irradiated data are quite different at the same conditions (see Fig. 2). Therefore, it is not a surprising finding that models fitted from fresh fuel data did not accurately predict the irradiated fuel data.

Irradiation adds complexity to fuel conductivity through the generation of defects in fuels, and modeling its effect with a few coefficients may not be sufficient. The models given in FRAPCON are only valid with the ranges of data used to fit the coefficients, and extrapolation beyond that range is questionable. The objective of this study was to extend the range of applicability by considering all data available (at the time of this study). There is also some uncertainty in the open literature data; however, that uncertainty does not sufficiently address the issue of why the data sets present such disparate results. Additional difficulties come from the fact that irradiation conditions are mostly calculations and not measurements of conductivity of irradiated samples, and the assumptions built into these calculations may be inconsistent with the conditions of the fuel after it is taken from the reactor or irradiation facilities.

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